

Anisotropy of the Superconducting State in Sr_2RuO_4

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The superconducting state emerges due to the formation and condensation of carrier Cooper pairs, although the exact microscopic mechanism responsible for the pairing in different materials varies and, in many cases, remains elusive. An important step towards a microscopic understanding is detailed knowledge of the superconducting order parameter. This is especially important in materials such as Sr_2RuO_4 (1, 2) and UPt_3 (3) where the carriers in the Cooper pairs are believed to form a triplet, introducing an additional degree of freedom and complexity to the problem. As a result the exact nature of the order parameter in these compounds, which have become paradigms for unconventional superconductivity, is still unresolved. We have studied the vortex lattice in Sr_2RuO_4 to measure the intrinsic anisotropy of the superconducting state between the c axis and the Ru-O basal plane ($\sim 60^\circ$), and found

this to greatly exceed the anisotropy of the upper critical field (~ 20). Our result imposes significant constraints on possible models of triplet pairing in Sr_2RuO_4 .

Introduction Multiple experimental and theoretical studies provide compelling support for triplet pairing of carriers (electrons and/or holes) and an odd-parity, p -wave order parameter symmetry in superconducting Sr_2RuO_4 . However, seemingly contradictory experimental results have left important unresolved questions concerning the detailed structure and coupling of the orbital and spin parts of the order parameter. A prominent example of this predicament is conflicting evidence as to whether the p -wave order parameter is chiral (4, 5).

In this Report we focus on the anisotropy of the superconducting state of Sr_2RuO_4 . The Fermi surface in this material consists of three largely 2-dimensional sheets with Fermi velocity anisotropies ranging from 57 to 174 (1, 6), and one would thus expect an upper critical field anisotropy within this range (7, 8). Experiments, however, find a much smaller upper critical field (H_{c2}) anisotropy $\Gamma_{H_{c2}} = H_{c2}^{\perp c} / H_{c2}^{\parallel c} \simeq 20$ and a near constant H_{c2} when the applied field is within $\pm 2^\circ$ of the basal plane (9). Within the same angular range the superconducting transition at H_{c2} becomes first-order, leading to suggestions of a subtle coupling between the magnetic field and the triplet order parameter (10), or Pauli limiting which is inconsistent with a Cooper pair zero spin-projection along the c axis (11).

We used small-angle neutron scattering (SANS) studies of the vortex lattice (VL) to determine the intrinsic anisotropy (Γ_{ac}) of the superconducting state in Sr_2RuO_4 yielding $\Gamma_{ac} \approx 60$. This is three times greater than $\Gamma_{H_{c2}}$, and poses significant constraints on the possible order parameter symmetry in this compound.

Materials and Methods The experiment was performed using a single crystal of Sr_2RuO_4 grown by the floating zone method and carefully annealed, yielding a critical temperature $T_c =$

1.45 K and no indication of a 3 K-phase (*I*). Measurements were performed at $T = 40 - 60$ mK using a dilution refrigerator (DR) inserted into a horizontal-field cryomagnet. Magnetic fields of $\mu_0 H = 0.5$ T and 0.7 T were applied close to the sample *a* axis. A motorized Ω -stage could rotate the DR within the magnet, allowing in-situ sample alignment and measurements as the crystalline basal plane was rotated with respect to **H**. A schematic of the experimental configuration is shown in Fig. 1(A). The VL was prepared by changing *H* and Ω at the base temperature, followed by a damped small-amplitude field modulation. This method produces a well-ordered VL and eliminates the need for a field-cooling procedure before each measurement. The SANS experiment was carried out on the D11 and D22 instruments at Institut Laue-Langevin (ILL), using a neutron wavelength $\lambda_n = 1.7$ nm and a wavelength spread $\Delta\lambda_n/\lambda_n = 10\%$. Some measurements were performed using polarized incident neutrons and a ^3He analysis cell to allow discrimination between spin-flip and non spin-flip scattering.

Results The VL scattered intensity in a SANS experiment is determined by the amplitude of the field modulation and is proportional to $|\mathbf{h}|^2$, where $\mathbf{h}(\mathbf{q})$ is the Fourier transform of the magnetic field $\mathbf{B}(\mathbf{r})$ (*12*). Using state-of-the-art SANS instruments at a high-flux neutron source such as ILL, it is possible to measure the diffraction from a well-ordered VL with a longitudinal Fourier coefficient $|h_z|$ as low as 0.1 – 1 mT, depending on the amount of background scattering (*13*). Here $|h_z| \propto \lambda_\perp^{-2}$, where λ_\perp is the average penetration depth in the screening current plane perpendicular to the applied field. Previous SANS studies with **H** \parallel **c** found a VL form factor for Sr_2RuO_4 no greater than a few mT (*14*). This indicates that measurements with **H** \perp **c** should not be possible as $|h_z^\perp|^2/|h_z^\parallel|^2 \propto (\lambda_{ab}/\lambda_c)^2 = \Gamma_{ac}^{-2}$, and with $\Gamma_{ac} \geq 20$ we estimate $|h_z^\perp| \leq 3 \mu\text{T}$, at least two orders of magnitude below what is required for a VL SANS experiment. However, in highly anisotropic superconductors such as Sr_2RuO_4 , there is a strong preference for the vortex screening currents to run within the basal *ab*-plane. As a result,

a small angle Ω between the applied field and the basal plane will lead to a significant transverse Fourier coefficient (h_x). Estimates based on an extended London model which includes an effective mass anisotropy yields $|h_x/h_z|^2 \propto \Gamma_{ac}^2$ (15), and thus predict $h_x^{\perp c}$ to be comparable in magnitude to $h_z^{\parallel c}$. As a result, scattering due to the transverse field modulation should be observable. This is confirmed by the VL diffraction pattern shown in Fig. 1(B) which show Bragg peaks aligned with the crystalline b direction (y axis), is in contrast to the exotic VL configurations predicted by Agterberg (16) and Kita (17).

Scattering from the transverse field modulation leads to a flip of the neutron spin ($\sigma \perp h_x$) and a Zeeman-splitting of the VL rocking curves shown in Fig. 2(A). Two maxima are observed for both the top (positive Q_y in Fig. 1(B)) and bottom (negative Q_y) VL reflection, as the angle (φ) between the scattering vector \mathbf{Q} and the direction of the incident neutron beam is varied to satisfy the Bragg condition (18). As expected, no scattering from the otherwise more commonly observed longitudinal VL field modulation (h_z) could be measured in Sr_2RuO_4 .

To verify that the observed diffraction is due to spin-flip scattering, measurements with a polarized neutron beam were performed. Here only one maximum is observed for each Bragg reflection, selected according to the direction of the neutron spin (18). Furthermore, the scattered intensity normalized to the incident neutron flux is doubled relative to the unpolarized beam as expected. Moreover, using polarization analysis it is possible to measure only the spin-flip scattering as shown in Fig. 1(B).

Dividing the integrated intensity by the incident neutron flux yields the integrated VL reflectivity

$$R = \frac{2\pi\gamma^2\lambda_n^2 t}{16\Phi_0 Q} |h_x|^2, \quad (1)$$

where $\gamma = 1.913$ is the neutron magnetic moment in nuclear magnetons, t is the sample thickness and $\Phi_0 = h/2e = 2069 \text{ T nm}^2$ is the flux quantum (13). As shown in Fig. 2 each peak is fitted to the sum of three Gaussians due to the asymmetry of the rocking curves (19). Moreover,

the integrated intensity for the two maxima (top, bottom) for a given reflection are added, as each corresponds to half the incident flux (one direction of the neutron spin). The form factor obtained in this fashion is shown in Fig. 3, for all measured fields and Ω 's. This illustrates how the VL SANS measurements are possible within a narrow angular range, with \mathbf{H} close to, but not perfectly aligned with, the basal plane. The width of the measurement “window” decreases with increasing field due to the rapidly decreasing $H_{c2}(\Omega)$ (9). In addition, the overall form factor decreases with increasing field. While the anisotropic London model provides a qualitative description of the enhanced field modulation (15), it is not able to provide a good quantitative fit to the data. As shown in Fig. 3, an extended London model that include a so-called core correction by multiplying the calculated $|h_x|$ by $\exp(-c Q^2(\Omega) \xi_{ab}^2)$ does still not yield a good fit to the data. Here $c \approx 1$, $Q(\Omega)$ is the magnitude of the VL scattering vector (see below) and $\xi_{ab} = (\Phi_0/2\pi H_{c2}^{\parallel c})^{1/2}$ is the in-plane coherence length (13). A quantitatively accurate model for the VL form factor is highly desirable as it would allow a determination of both λ and ξ .

We now turn to the main result of this Report, which is the measurement of the VL anisotropy. In an anisotropic superconductor the VL Bragg peaks are expected to lie on an ellipse with a major-to-minor ratio given by (7)

$$\Gamma_{VL} = \frac{\Gamma_{ac}}{\sqrt{\cos^2 \Omega + (\Gamma_{ac} \sin \Omega)^2}} \quad (2)$$

as shown in Fig. 4(A). This Ω -dependence was derived for anisotropic (but still 3-dimensional) superconductors, and was verified in early VL SANS measurements on $2H\text{-NbSe}_2$ with $\Gamma_{ac} = 3.2$ (20). Although Sr_2RuO_4 is a layered material, the coherence length along the c axis $\xi_c = 3.3$ nm is still several times greater than the Ru-O interlayer spacing (0.64 nm) (1), and we expect Eq. (2) to be applicable (21).

Due to the large anisotropy in Sr_2RuO_4 , VL Bragg peaks which are not on the vertical axis have scattering vectors essentially parallel to h_x making them unmeasurable as only compo-

nents of the magnetization perpendicular to \mathbf{Q} will give rise to scattering (22). Instead we determine the VL anisotropy based on flux quantization. Assuming that each vortex carries one flux quantum Φ_0 , the area of the reciprocal space ellipse in Fig. 4(A) is determined uniquely by the applied magnetic field. This yields $\Gamma_{VL} = (Q_0/Q)^2$, where Q is the magnitude of the measured VL scattering vector and $Q_0 = 2\pi(2\mu_0 H/\sqrt{3}\Phi_0)^{1/2}$ corresponds to an undistorted hexagonal VL ($\Gamma_{ac} = 1$). The magnitude of Q can be determined either from the position of the VL Bragg peaks on the detector as shown in Fig. 1(B), or from the peak positions $\varphi_1, \dots, \varphi_4$ in Fig. 2(A) (18). The two methods yield nearly identical results and using the average Q we obtain $\Gamma_{VL}(\Omega)$ shown in Fig. 4(B). Within the scatter in the data the results for both fields collapse onto a single curve, increasing upon approaching the a axis and reaching a value slightly above 50 before the intensity vanishes. If one assumes a quantization of $\Phi_0/2$, as recently reported for mesoscopic rings of Sr_2RuO_4 (23), the deduced values for Γ_{VL} would double. However, we consider this an unrealistic scenario in the present case, with a macroscopic, uniform sample. Fitting the data to Eq. (2) yields $\Gamma_{ac} = 58.5 \pm 2.3$. Only for angles within $\pm 1.3^\circ$ does the measured anisotropy deviate from that expected for an infinite ac anisotropy. Also shown for comparison is Γ_{VL} expected from the low temperature $\Gamma_{Hc2} = 20$ (9). We note that Γ_{ac} coincides with the anisotropy of the β Fermi surface sheet (57) (1, 6).

Discussion The large difference between Γ_{Hc2} and the intrinsic anisotropy of the superconducting state deep within the mixed phase measured by Γ_{ac} , indicates a strong suppression of the upper critical field in Sr_2RuO_4 for $\mathbf{H} \perp \mathbf{c}$. One possible explanation for this difference is Pauli limiting due to the Zeeman splitting of spin-up and spin-down carrier states by the applied magnetic field and the resulting reduction of the superconducting condensation energy (24). In spin-triplet superconductors the order parameter is most conveniently described in terms of the d -vector, directed along the zero spin projection axis where the configuration of the Cooper

pairs is given by $1/\sqrt{2}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$ (1, 2, 5). Consequently, Pauli limiting in the triplet case can only occur when $\mathbf{H} \parallel \mathbf{d}$. This is inconsistent with the chiral superconducting state proposed for Sr_2RuO_4 which requires $\mathbf{d} \parallel \mathbf{c}$ (2, 5), at least for the current experimental configuration with $\mathbf{H} \perp \mathbf{c}$.

Instead our results indicate that the superconducting state in this material should be understood based on either a subtle coupling between the magnetic field and the triplet order parameter (10); recent multiband p -wave models (25); a field-dependent mixing of singlet and triplet states (26) or singlet superconductivity (11). It should be noted however, that the latter (singlet pairing) does not provide an satisfactory explanation for the extreme sensitivity of T_c to impurities or to the chiral properties of Sr_2RuO_4 (1, 2).

Conclusion In conclusion, we have used SANS to measure the anisotropy of the superconducting state in Sr_2RuO_4 which greatly exceeds that of the upper critical field. This result imposes significant constraints on the possible pairing of carriers in this material. Any model aimed at describing the superconducting phase must provide a satisfactory explanation for this observation as well as the temperature dependence of $\Gamma_{H_{c2}}$, which increases with T and reaches Γ_{ac} at T_c (27), and the first-order nature of H_{c2} close to the basal plane (10).

References and Notes

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28. We acknowledge discussions with W. P. Halperin, V. G. Kogan and J. A. Sauls, and assistance with sample alignment by G. Sigmon. Research support was provided by the U.S. Department of Energy, Office of Basic Energy Sciences under Award DE-FG02-10ER46783 and by the MEXT of Japan KAKENHI No. 22103002.

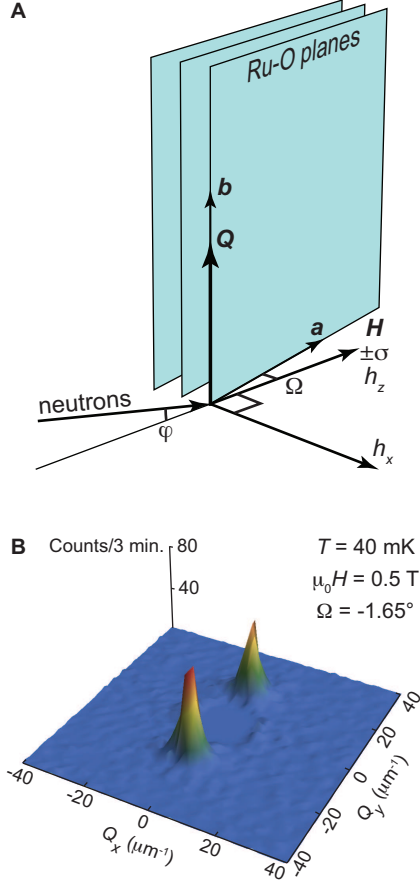


Figure 1: (A) Experimental geometry. The coordinate system is defined with z along \mathbf{H} and y in the Ru-O basal plane (along \mathbf{b}). The applied magnetic field \mathbf{H} is rotated away from the Ru-O (spanned by \mathbf{a} and \mathbf{b}) by an angle Ω . Neutron spins (σ) are parallel or antiparallel to the magnetic field. The incident neutron beam is in the yz -plane, at an angle φ relative to the field direction. The observed VL scattering vector is denoted \mathbf{Q} and the longitudinal and transverse component of the field modulation by h_z and h_x respectively. (B) Diffraction pattern showing spin flip scattering from the VL due to the transverse field modulation (h_x). The two Bragg peaks corresponds to $\pm\mathbf{Q}$ in the panel above. No background subtraction was performed but a small remnant of the undiffracted beam (due to the finite flipping ratio ~ 8) close to $Q = 0$ is masked off.

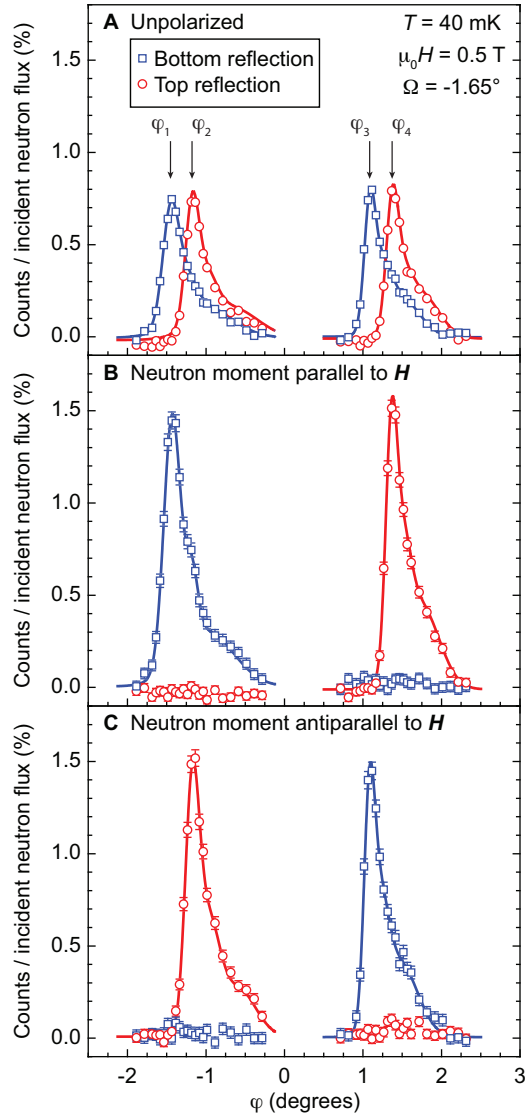


Figure 2: Vortex lattice rocking curves showing the scattered intensity as a function of the angle φ , for an unpolarized neutron beam (A) and for neutrons polarized with their magnetic moment parallel (B) and antiparallel (C) to the applied field. Except where shown, error bars are equal to or smaller than the symbols. With unpolarized neutrons two maxima are observed for both the bottom ($\varphi_{1/3}$) and top ($\varphi_{2/4}$) VL Bragg reflections. In all cases the intensity was normalized to the incident neutron flux. The curves are fits to the data as described in the text.

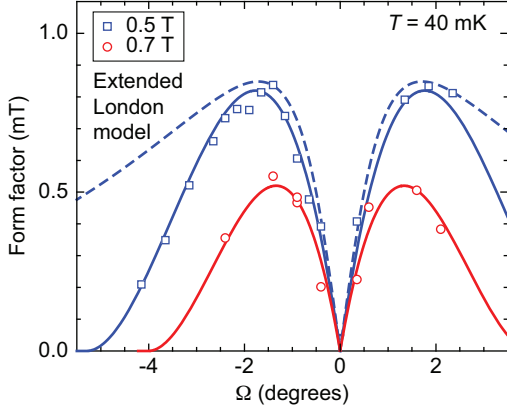


Figure 3: Vortex lattice form factor at 40 mK as a function of applied field and angle Ω with the a axis. The statistical error is roughly the size of the symbols. The solid lines are guides to the eye. The dashed line show an extended London model fit to the 0.5 T data as discussed in the text, with $\lambda_{ab} = 650$ nm, $\xi_{ab} = 66$ nm, $c = 1/2$ and $\Gamma_{ac} = 58.5$.

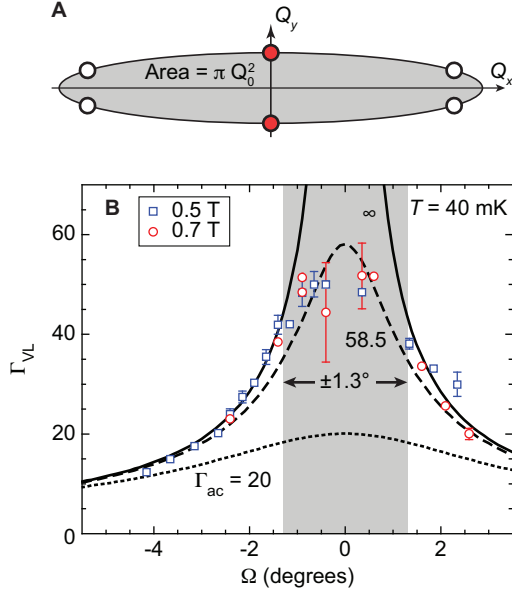


Figure 4: Vortex lattice anisotropy. (A) Schematic of VL Bragg reflections lying on an ellipse with major-to-minor axis ratio, $\Gamma_{VL} = 6$. Only the filled (red) peaks are observed. The reciprocal space area of the ellipse is $\pi Q_0^2 = 8\pi^3 \mu_0 H / \sqrt{3} \Phi_0$. (B) Measured VL anisotropy at 40 mK as a function of applied field and angle with the a axis (Ω). Except where shown explicitly the statistical error is the size of the symbols. The lines show the VL anisotropy calculated using Eq. (2) and $\Gamma_{ac} = 20$ (dotted), 58.5 (dashed) and ∞ (full).

Supplementary online material: Anisotropy of the Superconducting State in Sr_2RuO_4

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The two different directions of the neutron spin with respect to the applied field correspond to different nuclear Zeeman energies and lead to opposite shifts of the neutron momentum vector

$$k_{\uparrow(\downarrow)} = k_0 \sqrt{1 \pm \Delta\varepsilon/\varepsilon_0}, \quad (1)$$

where the subscript in parentheses henceforth correspond to the lower (minus) sign in the $\pm\Delta\varepsilon$ term. Here the nominal neutron wavevector $k_0 = 2\pi/\lambda_n$, $\varepsilon_0 = \hbar^2 k_0^2 / 2m_n$, $\Delta\varepsilon = \gamma\mu_N B$ and $\gamma = 1.913$ is the neutron magnetic moment in nuclear magnetons $\mu_N = e\hbar/2m_n = 31.5$ neV/T. With $\lambda_n = 1.7$ nm and $\mu_0 H = 0.5$ T one finds $k_{\uparrow}^2 - k_{\downarrow}^2 = 2 \times 10^{-4} k_0^2$.

Due to the short Q in the range $0.003 - 0.01 k_0$, the small shift in the neutron wavevector nonetheless leads to a significant difference in the angle ($\varphi_{1(2)}$) required to satisfy the Bragg

condition as shown in Fig. S2. In this case Bragg's law is replaced by

$$k_{\uparrow}^2 - k_{\downarrow}^2 \pm Q^2 = 2k_{\uparrow(\downarrow)} Q \sin \varphi_{1(2)}. \quad (2)$$

In contrast the scattering angles $\theta_1/\theta_2 = k_{\downarrow}/k_{\uparrow}$ are essentially identical for the two scattering configurations.

The magnitude of Q can be determined from the peak positions $\varphi_1, \dots, \varphi_4$ (see Fig. 2(A) in the main text) by

$$Q_{1(2)} = \mp k_{\uparrow(\downarrow)} \sin \varphi_{1(2)} \mp \sqrt{k_{\downarrow(\uparrow)}^2 - k_{\uparrow(\downarrow)}^2 \cos^2 \varphi_{1(2)}} \quad (3)$$

$$Q_{3(4)} = \mp k_{\downarrow(\uparrow)} \sin \varphi_{3(4)} \pm \sqrt{k_{\uparrow(\downarrow)}^2 - k_{\downarrow(\uparrow)}^2 \cos^2 \varphi_{3(4)}}, \quad (4)$$

using the same notation as earlier.

A more detailed discussion of the spin-flip scattering can be found in Ref. (1), where a similar but less extreme effect was observed in YBCO.

References and Notes

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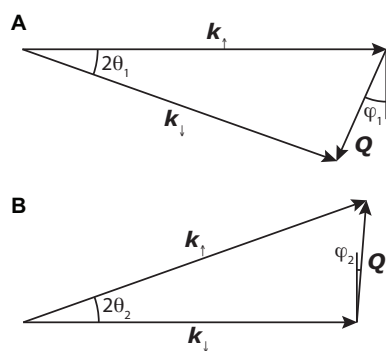


Figure S1: Schematics showing the scattering geometries corresponding to the reflection at $\varphi = \varphi_1$ (A) and φ_2 (B). Not to scale.